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Drying of a Liquid Droplet Suspended in a Binary Atmosphere

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ABSTRACT

In this work, we consider the spray drying of colloidal solutions, as applied to the case of the generation of synroc precursor powders. In this paper, a model of this process is formulated by considering the simplified situation of the evaporation of a pure liquid droplet suspended in a binary atmosphere consisting of the vapour phase of the liquid and an atmospheric gas such as air. Mass, momentum, and energy balances are given for the liquid and gas phases of the problem, and a binary diffusion equation is also given. The subsequent equation set is solved numerically, and the results are compared with experimental data, both of pure droplets, and of sol droplets.

NOMENCLATURE

C	concentration
\hat{C}	specific heat
D	mass diffusivity
f	external force
H	enthalpy
M	momentum transfer
p	pressure
Q	energy transfer
r	radial coordinate
\hat{r}	unit radial vector
R	droplet radius
\bar{R}	universal gas constant
t	time
T	temperature
u	velocity
U	thermal energy
w	mass fraction
x	transformed spatial coordinate
δ	unit tensor
κ	dimensionless parameter
λ	thermal diffusivity
π	pressure tensor
ρ	density
σ	surface tension

Subscripts

a	atmosphere
E	“east” node for computation
g	gas
i	either vapour (v) or atmosphere (a)
j	gas (g), vapour (v), or atmosphere (a)
l	liquid
P	constant pressure OR central node for computation
v	vapour
V	constant volume

INTRODUCTION

The production of the synthetic rock known as synroc (Sizgek, Bartlett and Brungs 1998), a substance used for encapsulating high level liquid nuclear waste, requires a number of stages. The first stage involves the preparation of a precursor Titanium, Zirconium, Aluminium (TZA) colloidal solution, or sol (Sizgek, *et al.* 1998). Once the TZA sol is prepared, it is injected into a spray drying chamber so that a fine TZA powder is produced. However, depending on the properties of the sol and the conditions of the chamber, the resulting powder can have different morphologies. The most desirable of these is a solid, porous sphere. Some examples of undesirable morphologies are hollow spheres and tori. The morphology of the powder can have a significant impact on the effectiveness of the downstream synroc product to encapsulate nuclear waste (Sizgek, *et al.* 1998). In this paper we present and validate an initial model for the spray drying process in which we consider a liquid droplet evaporating in an atmosphere consisting of its own vapour and an inert atmospheric gas such as air or argon.

We consider the spray drying process for sols to consist of four distinct phases. First, the atomised colloidal solution is released into the drying chamber, during which rapid transient behaviour occurs. Following this transient period, the droplets evaporate until colloid at the surface solidifies to form a crust. Next, liquid is evaporated through the pores of the crust; this causes the crust to thicken. Finally, when all of the colloid particles have solidified, the remaining water is evaporated off through heating until the powder exits the drying chamber.

From a simplified perspective, the spray drying process can be considered to be the process of the evaporation of a single droplet in an infinite atmosphere under the action of gravity. A further simplification involves neglecting gravity and the assumption of a stagnant atmosphere. This results in a model of the process that is radially symmetric, thereby permitting a one-dimensional analysis.

A number of simplified models of spray drying have been considered in the literature. Sirignano (1999) considers the process as it applies to fuel combustion. He formulates quite complex models of the process; however, certain important concepts in fluids and evaporation, such as surface tension and vapour pressure, are not considered. This may be an acceptable approach for combustion models, but for inert drying, this is not valid. Van der Lijn (1976) models the spray drying of liquid foodstuffs (such as milk). In doing so, however, he makes numerous simplifications, such as neglecting the thermal

distributions in the droplet. Sano and Keey (1982) attempt to model the formation of hollow sphere morphologies during the spray drying of sol droplets. However, they make the *a priori* assumption that a bubble will form inside the droplet, yet they do not provide proof that this is the mechanism by which hollow spheres are formed.

Oberman, Farrell and Sizgek (2005) developed a model that considered the evaporation of a liquid droplet in its own, pure atmosphere. The primary purpose of constructing the model was to create a numerical framework that could be adapted for more complex models. The most notable use for such a framework is for the contemplation and testing of methods by which the moving boundary between the liquid and gas phases can be managed numerically. Oberman *et al.* assumed that the liquid phase is incompressible and pure, that the gas phase is ideal, inviscid, and infinite, and that it is the vapour form of the liquid phase. It is also assumed that there are no external forces such as gravity and that both phases are initially in internal equilibrium. In addition, the system is assumed to be spherically symmetric, and the gas phase is assumed to be far less dense than the liquid phase. A regular perturbation analysis was carried out on the model equations and this led to the assumption that the vapour phase is isobaric. Boundary fixing transformations were applied to the liquid and gas phases of the problem and the resulting model equations were solved numerically using control volume (Patankar 1980) discretisation methods. The model simulations were found to agree favourably with the well-known “d²-law” (Nomura, Ujiie, Rath, Sato and Kono 1996); however, due to a lack of relevant experimental data, Oberman *et al.* do not validate their results.

In this paper, we adopt an approach similar to that of our previous work (Oberman, *et al.* 2005) in that we again consider a single droplet, containing homogeneous liquid, but not colloid, that is being dried in a quiescent, inert atmosphere. Unlike the previous work, however, we now consider this atmosphere to be binary in nature; that is, it consists of an inert atmospheric gas and the vapour phase of the liquid. This model will therefore capture the essence of the first two stages in the drying process described above. The purpose of this model is to construct a system of equations for the gas phase that will then be implemented in a more complex model involving colloidal behaviour. It also permits the confirmation of the model design by comparison with experimental data, and the results of such a validation process are included here.

MODEL DEVELOPMENT

The following simplifying assumptions will be applied to this model:

1. The liquid phase is incompressible and pure.
2. The gas phase is an ideal mixture of atmospheric gas and vapour, each of which is ideal, inviscid, and infinite.
3. There are no outside forces acting upon the system.
4. The entire system is spherically symmetric.
5. The gas phase has significantly lower density than the liquid phase.
6. Initially, each phase is in internal equilibrium.
7. The atmospheric gas does not dissolve into the liquid phase.

Each assumption has a basis in logic; Assumptions 1, 3, and 6 are due to the physical setup of the model, Assumptions 2, 4, and 7 simplify the system without oversimplifying the underlying physics, and Assumption 5 is essential for realistic thermodynamic behaviour, although it is often not explicitly stated.

As a result of the above assumptions, the following model equations can be formulated. We note that unless otherwise stated, all quantities introduced in the following equations are dimensionless.

Inside the droplet

Within the droplet, the behaviour of the liquid essentially remains the same as in our earlier model. Although equations for mass, momentum, and energy balance may be derived for the liquid phase, the mass and momentum equations can be reduced via Assumption 1. This reduction leads to the conclusion that the radial velocity of the liquid is zero, and also that the pressure is independent of position within the liquid.

The remaining equation, namely the energy balance equation, takes the form (Bird, Stewart and Lightfoot 1960)

$$\frac{\partial T_l}{\partial t} = \frac{\kappa_l}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial T_l}{\partial r} \right). \quad (1)$$

We consider that the initial temperature of the droplet is uniform and is given as $T_i = 1$. Furthermore, at the centre of the droplet, symmetry requires that

$$\frac{\partial T_l}{\partial r} = 0. \quad (2)$$

Outside the Droplet

The gas phase of the binary gas model consists of two subphases, namely the vapour (v) and atmosphere (a) subphases. The vapour subphase consists of vaporised liquid, while the atmosphere subphase consists of those gases not in the vapour subphase.

Noting Assumption 2 we may assert that, in the gas phase, pressure is additive, giving

$$p_g = p_v + p_a. \quad (3)$$

In addition, for each of these the ideal gas law applies, namely (Roberts 1994),

$$p_j = C_j \bar{R} T. \quad (4)$$

Noting Assumption 2, the continuity equation takes the form (Bird, *et al.* 1960),

$$\frac{\partial \rho_j}{\partial t} = -\frac{1}{r^2} \frac{\partial}{\partial r} (r^2 u_j \rho_j), \quad (5)$$

where the velocity scaling has been chosen to set the dimensionless parameter in this equation to 1. In order for this equation to be satisfied for both the gas phase and its subphases, it is necessary to define the velocity of the gas to be the mass average velocity (Bird, *et al.* 1960), namely,

$$\rho_g u_g = \rho_v u_v + \rho_a u_a. \quad (6)$$

Alternatively, using the mass fractions, $w_v = \rho_v/\rho_g$, and $w_a = 1 - w_v$, we may write Equation (6) as

$$u_g = w_v u_v + w_a u_a. \quad (7)$$

Additionally, we may write the subphase velocities in terms of the mass average velocity and subphase mass fractions, namely (Bird, *et al.* 1960),

$$\rho_i u_i = \rho_i u_g - \kappa_2 \rho_g \frac{\partial w_i}{\partial r}. \quad (8)$$

The first term on the right of this expression represents an advective flux and the second term represents the mass flux associated with Fickian diffusion.

The momentum balance equations for each subphase (v or a) can be written, in *dimensioned* form, as (Bird, *et al.* 1960)

$$\begin{aligned} \frac{\partial \rho_i u_i}{\partial t} + \frac{1}{r^2} \frac{\partial}{\partial r} (r^2 \rho_i u_i^2) \\ = \rho_i f_i + \hat{\mathbf{r}} \cdot (\nabla \cdot \boldsymbol{\pi}_i) + M_i. \end{aligned} \quad (9)$$

Summing this equation over both subphases and applying Equation (6) and Assumption 3 gives

$$\begin{aligned} \frac{\partial \rho_g u_g}{\partial t} + \frac{1}{r^2} \frac{\partial}{\partial r} (r^2 (\rho_v u_v^2 + \rho_a u_a^2)) \\ = \hat{\mathbf{r}} \cdot (\nabla \cdot \boldsymbol{\pi}_g), \end{aligned} \quad (10)$$

where $M_v + M_a$ is zero because it represents momentum transfer from one subphase to the other, which, of course, must be conserved. In addition, $\boldsymbol{\pi}_g$ can be reduced, using Assumption 2, to the form $-p_g \hat{\mathbf{d}}$. This allows Equation (10) to be written as,

$$\begin{aligned} \frac{\partial \rho_g u_g}{\partial t} + \frac{1}{r^2} \frac{\partial}{\partial r} (r^2 (\rho_v u_v^2 + \rho_a u_a^2)) \\ = -\frac{\partial p_g}{\partial r}. \end{aligned} \quad (11)$$

In turn, Equation (11) can be simplified, and using Equations (3), (6), and (8) and the definition of material derivative (Roberts 1994) to arrive at the final form of the gas phase momentum balance equation, which upon the introduction of dimensionless variables is given by,

$$\begin{aligned} \frac{\partial \rho_g u_g}{\partial t} + \frac{1}{r^2} \frac{\partial}{\partial r} (r^2 \rho_g u_g^2) \\ = -\kappa_3 \frac{\partial p_g}{\partial r} - \frac{\kappa_2^2}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\rho_g}{w_v w_a} \left(\frac{\partial w_v}{\partial r} \right)^2 \right). \end{aligned} \quad (12)$$

The total energy balance equations for each subphase can be written, in *dimensioned* form, as (Bird, *et al.* 1960)

$$\begin{aligned} \frac{\partial}{\partial t} \left(\rho_i \left(U_i + \frac{u_i^2}{2} \right) \right) + \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \rho_i \left(U_i + \frac{u_i^2}{2} \right) u_i \right) \\ = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \left(\lambda_i \frac{\partial T}{\partial r} - p_i u_i \right) \right) + Q_i, \end{aligned} \quad (13)$$

Introducing the enthalpy, $H_i = U_i + p_i/\rho_i$ (Bird, *et al.* 1960), and simplifying, we obtain that

$$\begin{aligned} \frac{\partial}{\partial t} \left(\rho_i \left(H_i + \frac{u_i^2}{2} \right) \right) + \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \rho_i \left(H_i + \frac{u_i^2}{2} \right) u_i \right) \\ = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \lambda_i \frac{\partial T}{\partial r} \right) + \frac{\partial p_i}{\partial t} + Q_i. \end{aligned} \quad (14)$$

Summing across the subphases, we have

$$\begin{aligned} \frac{\partial}{\partial t} \left(\rho_v \left(H_v + \frac{u_v^2}{2} \right) + \rho_a \left(H_a + \frac{u_a^2}{2} \right) \right) + \\ \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \left[\rho_v \left(H_v + \frac{u_v^2}{2} \right) u_v + \rho_a \left(H_a + \frac{u_a^2}{2} \right) u_a \right] \right) \\ = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \lambda_g \frac{\partial T}{\partial r} \right) + \frac{\partial p_g}{\partial t}, \end{aligned} \quad (15)$$

where energy conservation requires that $Q_v + Q_a = 0$.

Letting $\rho_v H_v + \rho_a H_a = \rho_g H_g$ and applying equation (8) yields,

$$\begin{aligned} \frac{\partial}{\partial t} \left(\rho_g \left[H_g + \frac{u_g^2}{2} + \frac{D^2}{2w_v w_a} \left(\frac{\partial w_v}{\partial r} \right)^2 \right] \right) + \\ \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \rho_g \left[H_g + \frac{u_g^2}{2} + \frac{D^2}{2w_v w_a} \left(\frac{\partial w_v}{\partial r} \right)^2 \right] u_g \right) \\ = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \lambda_g \frac{\partial T}{\partial r} \right) + \frac{\partial p_g}{\partial t} + \\ \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \rho_g D \frac{\partial w_v}{\partial r} \left[H_v - H_a - \frac{DK}{w_v w_a} \right] \right), \end{aligned} \quad (16)$$

where K is given by,

$$K = u_g \frac{\partial w_v}{\partial r} - \frac{D}{2} \left(\frac{1}{w_v} - \frac{1}{w_a} \right) \left(\frac{\partial w_v}{\partial r} \right)^2. \quad (17)$$

Noting that $H = \hat{C}_p T$, $H = U + p/\rho$, and $U = \hat{C}_v T$, and introducing dimensionless parameters, we may write the above total energy equation in terms of the specific heats, namely,

$$\begin{aligned} \frac{\partial}{\partial t} \left(\rho_g \left[\hat{C}_{vg} T + \frac{u_g^2}{2} + \frac{\kappa_4}{w_v w_a} \left(\frac{\partial w_v}{\partial r} \right)^2 \right] \right) + \\ \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \rho_g \left[\hat{C}_{vg} T + \frac{u_g^2}{2} + \frac{\kappa_4}{w_v w_a} \left(\frac{\partial w_v}{\partial r} \right)^2 \right] u_g \right) = \\ \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \left[\left[\kappa_5 \lambda_g \frac{\partial T}{\partial r} - \kappa_6 p_g u_g + \right. \right. \right. \\ \left. \left. \left. \rho_g \frac{\partial w_v}{\partial r} \left[\kappa_7 T - \frac{\kappa_4}{w_v w_a} \frac{\partial w_v}{\partial r} \frac{\partial u_g}{\partial r} + \frac{\kappa_4}{2w_v w_a} \left(\frac{1}{w_v} - \frac{1}{w_a} \right) \left(\frac{\partial w_v}{\partial r} \right)^2 \right] \right] \right] \right), \end{aligned} \quad (18)$$

Initially, the density of the gas phase is uniformly $\rho_g = 1$, the radial velocity is uniformly $u_g = 0$, the temperature is uniformly $T = 1$, and the mass fraction is uniformly $w_v = w_{v0}$ (a constant). At large distances from the droplet, ρ_g , w_v , and T remain at these constant values.

Equations (5), (12), and (18), along with Equation (8) and the boundary conditions described above, govern the variables ρ_g , w_v , u_g , and T outside the liquid droplet.

On the Surface of the Droplet

The following equations apply only at the surface, $r = R(t)$, of the liquid droplet. Together they form a set of conditions that describe the conservation of mass, momentum and energy at the moving boundary between the liquid and gas as the radius of the droplet decreases due to evaporation (or increases due to condensation).

The liquid and gas phases are in thermal contact at the surface, and therefore continuity of temperature applies (Margerit and Sero-Guillaume 1996), namely,

$$T = \kappa_9 T_l \quad (19)$$

at the surface of the droplet, where the dimensionless parameter κ_9 appears due to the different nondimensionalisations applied to the two temperatures.

All thermal energy conducted into the droplet from the gas is used for either sensible heating of the droplet or to supply latent heat energy to the liquid being evaporated. As such, the heat flux surface condition can be formulated as (Delhaye 1981)

$$\lambda \frac{\partial T}{\partial r} = \kappa_{10} \frac{\partial T_l}{\partial r} - \kappa_{11} \dot{R}. \quad (20)$$

All mass evaporated from the liquid phase must necessarily enter the vapour subphase. Thus the conservation of mass flux at the interface yields (Delhaye 1981),

$$\rho_v(u_g - \dot{R}) - \kappa_2 \rho_g \frac{\partial w_v}{\partial r} = -\kappa_{12} \dot{R}. \quad (21)$$

Noting Assumption 7, the mass flux of atmosphere across the interface must be zero, and so, noting that $w_a = 1 - w_v$, we obtain (Delhaye 1981)

$$(1 - w_v)(u_g - \dot{R}) + \kappa_2 \frac{\partial w_v}{\partial r} = 0. \quad (22)$$

The pressure at the surface of the droplet is found by generalising the Clapeyron equation to unequal pressures. This gives (Lock 1994)

$$\frac{dp_l}{dt} - \frac{\kappa_{12}}{\rho_v} \frac{dp_v}{dt} = -\frac{\kappa_{13}}{T} \frac{dT}{dt}. \quad (23)$$

The difference between the gas and liquid pressures can be derived by considering a balance of forces, namely (Delhaye 1981),

$$p_g = p_l - \frac{\kappa_{14}}{R} + \kappa_{15} \dot{R} u_v. \quad (24)$$

Summary

Equation (1) with the boundary condition in Equation (2), Equation (5) with Equation (8), Equations (12) and (18), the related initial and infinity conditions, plus, at the interface, and Equations (19) through (24), form a complete system of equations describing the evaporation (or condensation) of a liquid droplet suspended in a binary atmosphere consisting of an ideal mixture of pure vapour and pure atmospheric gas.

Analysis

Of the dimensionless parameters introduced above, four are significant in terms of analysing the system of model equations. These are,

$$\begin{aligned} \kappa_2 &= \frac{Dt_0}{R_0^2} & \kappa_3 &= \frac{p_\infty t_0^2}{\rho_\infty R_0^2} \\ \kappa_4 &= \frac{D^2}{2R_0^2 \hat{C}_{v\infty} T_\infty} & \kappa_{14} &= \frac{\sigma_0}{p_\infty R_0} \end{aligned}$$

If we set κ_2 equal to one, then the time scale of the nondimensionalisation is that of the process of diffusion of water vapour into air. On this time scale, κ_3 is found to be exceptionally large, indicating rapid propagation of pressure. This fact and consideration of boundary conditions reveals that the pressure of the system can be considered isobaric. However, assumption of isobaric conditions leads to a numerically unstable system, and this result therefore is not used to simplify our system of equations. The parameter κ_4 represents the proportion of the energy attributable to the process of diffusion. It is negligible, and is taken to be zero in our equation system. Finally, κ_{14} represents the ratio of the surface tension and surface pressure. This is also found to be negligible, and is subsequently set to zero in our system.

NUMERICS

In order to solve the above system numerically, it is first necessary to decide upon a mesh scheme. The simplest method of handling the changing boundary is to lock the surface at $x=1$ by scaling the spatial coordinate within the droplet using $r = xR(t)$, and by translating the spatial coordinate in the gas phase using $r = x + R(t) - 1$ (Oberman, *et al.* 2005). Once this is done, a simple linear mesh is applied to the system.

The form of the equations seen above is conducive to the application of a Control Volume Scheme (Patankar 1980), with the exception of the κ_3 term in the momentum equation (Equation (12)). During discretisation, we find an integral of the form

$$\int_{x_p}^{x_E} (x + R(t))^2 \frac{\partial p}{\partial x} dx,$$

which, after applying the product rule and using an inverse-linear fit for p gives

$$r_E r_P (p_E - p_P).$$

In addition, we apply upwinding (Patankar 1980) to the advective terms and note that the velocity used for upwinding is that relative to the mesh, which takes the form, $u_g - \dot{R}$.

Once the equations are discretised, we obtain a system of nonlinear difference equations. In order to solve this system, we apply a fully-implicit iterative Newton method (Burden and Faires 1997), alternately solving within and without the droplet, and repeating until convergence for each time step is achieved. A computer code implementing this numerical scheme has been written in MATLAB.

RESULTS & DISCUSSION

This system differs from the previous homogeneous model of Oberman *et al.* (2005) due to the inclusion of a second, atmospheric gas within the gas phase. To see the effect of this, we compare the results of the code under a variety of conditions within the gas phase.

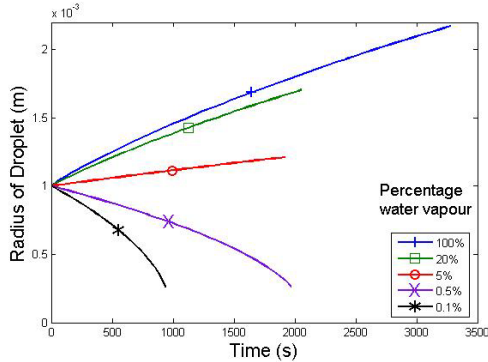


Figure 1: Comparison of droplet drying curves for varying humidity.

Figure 1 displays a comparison of the model for a droplet of radius 1 mm in an atmosphere at 28°C and 101.3 kPa. It can be seen that the homogeneous model (100% water vapour) predicts strong condensation due to the temperature being significantly below the dew point, while under very dry conditions (0.1% water vapour), where the gas phase has a very low mass fraction of water vapour, it is seen that evaporation occurs quite rapidly. Under more realistic conditions (0.5% water vapour), the gas phase temperature is only slightly above the dew point temperature and we see that evaporation is slow.

By examining the pressure distribution obtained numerically, we find that, on the time scale of evaporation, p_g only deviates by $10^{-7}\%$ from its initial value, indicating without doubt that the gas pressure can be considered isobaric. This is found to be true even under extreme conditions, where the initial temperatures are

greater than the boiling point of water and the atmosphere is almost completely dry. This outcome is consistent with our previous observation concerning the parameter κ_3 .

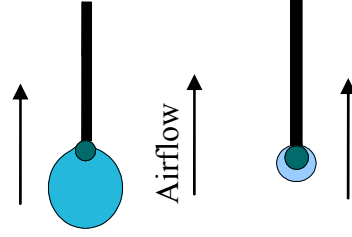


Figure 2: Experimental design imperfections.

In order to validate the results of this model, a number of experiments were performed. These experiments involved the evaporation of a droplet suspended on the end of a platinum wire (see Figure 2), which was then inserted into a thermogravimetry device. For the duration of the evaporation, the mass of the droplet was recorded. On the left side of Figure 2, we see a representation of a droplet at early times; it can be seen that the droplet is only slightly affected by the presence of the platinum wire. On the right side of Figure 2, we see that the droplet radius is no longer large compared to the thickness of the wire, and this artificially increases the surface area of the droplet.

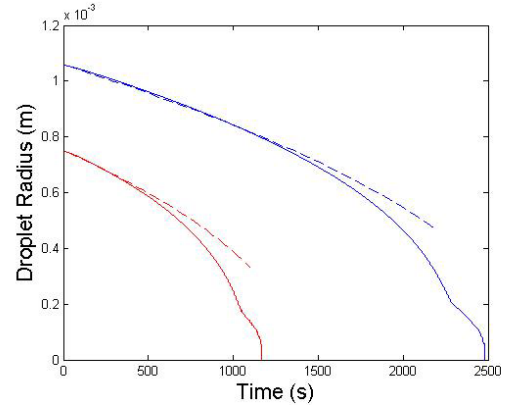


Figure 3: Comparison of experiment (—) and theory (---) for pure water droplets.

Figure 3 compares the experimental evaporation data for two pure water droplets of different initial radii with the numerical predictions. It can be seen that, in both cases, the numerics predict a slower evaporation rate at later times than is seen in the experiments. This can be attributed to three factors: the surface area of the droplet is artificially increased by the presence of the platinum wire; there is forced airflow within the apparatus, which reduces the humidity of the air; and there is increased heat transfer through the platinum wire. These three issues are not accounted for by the model.

Figure 4 compares the experimental evaporation data (solid lines) for water droplets of different initial radii containing colloidal materials with the corresponding numerical simulations (dashed lines) of our model. We note that even though our model does not account for the presence of colloidal material in droplets, the model

adequately predicts behaviour for early times, when the droplet still behaves similarly to a pure water droplet. At late times, the experimentally observed evaporation curves show a change in concavity due to the coagulation of colloid particles. Note that the spike in the experimental data at approximately 200s was an artefact of the measuring apparatus used.

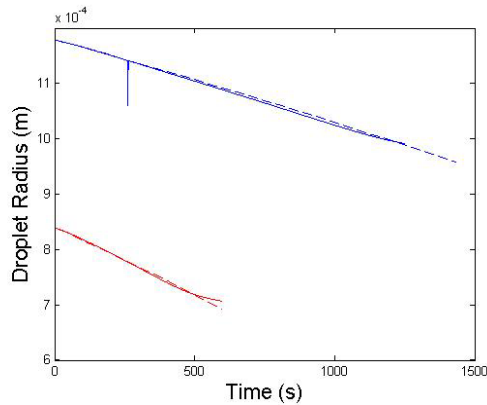


Figure 4: Comparison of experiment (—) and theory (---) for sol droplets.

CONCLUSION

A model of the evaporation of a pure liquid droplet suspended in a binary atmosphere has been developed. A numerical scheme has been developed to solve this system, and the scheme was implemented as a MATLAB code. The results of the code were compared to experimental data for various droplet radii, and they were found to favourably agree. Comparison with experimental data for the drying of sol droplets demonstrated further agreement, indicating that such a model is accurate for the first two stages of spray drying of synroc precursor sols, during the early stages when the colloidal material has yet to coagulate to any appreciable degree.

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APPENDIX A

$$\kappa_1 = \frac{Dt_0}{R_0^2}$$

$$\kappa_3 = \frac{p_\infty t_0^2}{\rho_\infty R_0^2}$$

$$\kappa_5 = \frac{\lambda_g t_0}{R_0^2 \rho_\infty \hat{C}_{V\infty}}$$

$$\kappa_7 = \frac{Dt_0(\hat{C}_{Pv} - \hat{C}_{Pa})}{R_0^2 \hat{C}_{V\infty}}$$

$$\kappa_9 = \frac{T_0}{T_\infty}$$

$$\kappa_{11} = \frac{L\rho_l R_0^2}{\lambda_g T_\infty t_0}$$

$$\kappa_{13} = \frac{L\rho_l}{p_\infty}$$

$$\kappa_{15} = \frac{\rho_l R_0^2}{p_\infty t_0^2}$$

$$\kappa_2 = \frac{Dt_0}{R_0^2}$$

$$\kappa_4 = \frac{D^2}{2R_0^2 \hat{C}_{V\infty} T_\infty}$$

$$\kappa_6 = \frac{p_\infty}{\rho_\infty \hat{C}_{V\infty} T_\infty}$$

$$\kappa_8 = \frac{D}{2t_0(\hat{C}_{Pv} - \hat{C}_{Pa})T_\infty}$$

$$\kappa_{10} = \frac{\lambda_l T_0}{\lambda_g T_\infty}$$

$$\kappa_{12} = \frac{\rho_l}{\rho_\infty}$$

$$\kappa_{14} = \frac{\sigma_0}{p_\infty R_0}$$